Ph.D. Thesis booklet

Graphene-based heterostructures under pressure

Bálint SZENTPÉTERI

Department of physics

Budapest University of Technology and Economics

2025

Supervisor: Péter MAKK
Associate Professor
Department of Physics
BUTE

1 Introduction

The continuous development of our technology always needs novel materials with the desired properties. With the gate all around field effect transistors, the conventional semiconducting industry in a few years is reaching its limit by reaching the 0.5 nm node[1]. To go beyond it, a fundamental change is necessary to be able to engineer the material on the level of atomic precision in the next generation of microelectronic devices[2]. An alternative approach became available with the discovery of two-dimensional (2D) materials[3] after the first isolation of graphene[4]. Among the 2D materials, besides graphene, which is a semimetal, there are insulators like hBN, semiconductors like transition metal dichalcogenides (TMDs), topological insulators and so on[5, 6].

What makes 2D crystals unique compared to conventional materials is that they can be placed on top of each other layer by layer with atomic precision in the vertical direction, realizing van der Waals (vdW) heterostructures[5, 7]. The properties of these heterostructures are determined by the nature of the constituting materials and the interlayer interactions[8, 9]. For example, hBN can be used as protective layers by encapsulating the graphene within hBN layers to protect it from the environment and improve the charge carrier mobility in it[10, 11]. In hBN/graphene heterostructures, the lattice mismatch may lead to the formation of a moiré pattern, which depends on the relative orientation of the crystals, the twist angle. A superlattice forms, where the moiré pattern acts as a periodic potential[12].

The twisting is a unique degree of freedom of vdW heterostructures, unlike conventional heterostructures. Twisting can fundamentally change the properties of a heterostructure, leading to new properties such as correlated phases and superconductivity in twisted bilayer graphene[13, 14]. The field of twistronics, which focuses on the effect of twisting is dynamically growing by including newer and newer systems such as twisted TMD heterostructures[15].

Besides twistronics, vdW materials are also exciting for the field of spintronics. Spintronics focuses on using the spin degree of freedom of the charge carriers to realize electronic devices, which are controlled with the manipulation of electron spins instead of the charge[16]. The manipulation is usually done by a magnetic field or with the help of spin-orbit coupling (SOC). Most spintronic devices are based on spin valves, which are heterostructures made of magnetic layers with a nonmagnetic layer between them. For instance, an insulator in a tunneling magnetoresistance device which is a ferromagnet/insulator/ferromagnet heterostructure can be used as a memory element in hard drives. Moreover, a non-magnetic metal is used in spin-transfer torque magnetic random-access memories, where the spin-polarized current is used to control the memory state[17]. These heterostructures can also be realized with 2D materials, where the graphene usually serves as a transport channel due to its excellent properties like negligible SOC and the near-absence of nuclear spins[18, 19]. Moreover, combining graphene with TMDs induces SOC in graphene with proximity effects[20], and the spin current in these heterostructures could be controlled with electrical gating[21–24] and with spin-to-charge conversion[25–28] bringing the 2D materials closer for industrial applications.

In vdW heterostructures, as the interlayer interactions play a crucial role, their properties could be altered by tuning the strength of these interactions[9]. Varying the interlayer distance could significantly change the strength of interlayer interactions, which could be done in practice by pressing the heterostructure[29]. Realizing novel phases with pressure could lead to new applications and are very important in fundamental research. Therefore, this thesis aims to investigate the effect of pressure on vdW heterostructures using electrical transport techniques.

Before this thesis, experimental research activity in the BME Nanoelectronics Lab focussed on the transport properties of single-layer graphene (SLG), placed on Si/SiO_2 substrate or suspended in vacuum[30–34]. After that, the effect of pressure is investigated on SLG-based heterostructures and other materials[35, 36].

2 Objectives

The study of the effect of pressure on the transport properties of vdW heterostructures is a relatively new field[35, 37–39]. My main aim is to extend this field, by performing hydrostatic pressure-dependent measurements on novel vdW heterostructures.

To observe new physics in graphene such as electron optics[40], Moiré physics[41], fractional quantum Hall effect[42] or highly viscous electron fluids[43, 44] and also for the industrial application of graphene, reliably high-quality sample fabrication is essential. To achieve this, it is essential to know the main source of disorder and the limiting factors of the mobility to be able to eliminate them during fabrication. By encapsulating SLG in hBN crystals, high-mobility devices can be realized[10, 45, 46]. In these heterostructures, the main source of disorder at low temperatures is long-ranged and at higher temperatures, the electron-phonon coupling starts to dominate it. One of the objectives of this work was to study these heterostructures under pressure to try to find out the origin of the disorder.

Graphene is an ideal material for spintronics due to its excellent properties, such as low spin-orbit coupling (SOC), and hyperfine coupling[19, 47]. However, to use graphene in spintronic devices, electrical control of the spin information is necessary[20, 48], for which large SOC is required. It was theoretically predicted that the SOC can be increased by bringing graphene in proximity to a transition metal dichalcogenide (TMD), which has a strong intrinsic SOC[20]. This was later demonstrated with experiments[21–23, 27, 28, 49–62]. My objective was to check whether the effect of pressure can further enhance the induced SOC, or how it influences the system. My tasks involved experimental and theoretical works.

Among the twisted structures, the twisted double bilayer graphene (TDBG) is very interesting, as it is tunable with electric field and it exhibits correlated insulator and topologically non-trivial phases[63–71]. In twisted structures, the interlayer interactions play a crucial role, which can be tuned with pressure. My objective was to verify this and check the effect of pressure on the band structure both experimentally and theoretically.

3 New scientific results

The results of my thesis are summarized in the following thesis points:

1. **I showed for the first time the extensive tunability of the moiré gaps of twisted double bilayer graphene with pressure in agreement with the theory.** I performed temperature and bias voltage-dependent transport experiments. From the thermal activation and bias voltage-dependent measurements, I measured the moiré gaps of a TDBG near the magic angle and showed that the moiré gaps in a TDBG can be decreased and fully closed by applying hydrostatic pressure. Furthermore, I showed by measuring Brown-Zak oscillations that, the twist angle doesn't change with pressure. Finally, I also observed a decrease in the correlation effects by increasing the pressure. [T1]

2. **I showed for the first time in WSe_2/BLG heterostructures the enhancement of the proximity-induced Rashba-type and Ising-type SOC with hydrostatic pressure in agreement with the theory.** I made low-temperature magnetic field-dependent experiments on WSe_2/BLG heterostructures. In the experiments, I used the Shubnikov-de Haas oscillations to obtain the Rashba-type SOC strength, and I used the quantum Hall effect to obtain the Ising-type SOC strength by measuring the positions of the Landau level crossings. I showed that with pressure, in WSe_2/BLG heterostructures, the positions of the crossing points change and also I showed that the pressure increases the splitting of the Fermi surface, which is due to the lifted spin-degeneracy of the SOC. From these, I found that the proximity-induced spin-orbit coupling strength in WSe_2/BLG heterostructures increased by more than 50% under pressure. [T2]

3. **I showed the decrease and closing of the moiré gaps in twisted double**

bilayer graphene with pressure with simulations, which showed a good agreement with the experiments [T1]. I simulated the change of the Fermi surfaces of WSe_2/BLG heterostructures by varying the Rashba-type SOC and I calculated the change of the Landau level crossings by varying the Ising-type SOC [T2]. Using my simulations, I obtained the SOC strengths by fitting the experimental data with the models [T2, T3]. I successfully applied the Bistritzer-MacDonald model on twisted double bilayer graphene, where I used the pressure dependence of the interlayer tunneling from the literature to calculate the band structure at different pressures [T1]. I found that, by increasing the pressure, the moiré gaps decrease and fully close, which is in good agreement with my experiments [T1]. In the case of WSe_2/BLG heterostructures, I calculated band structure from a low-energy model [T2]. From the band structure, I calculated the Fermi surfaces at different Rashba-type coupling strengths. Then I fitted the experimentally obtained Fermi surfaces with the model and obtained the Rashba-type SOC strength [T2]. From the low-energy model of the heterostructure, I calculated numerically the Landau level energies as a function of an applied external electric field. I calculated the change of the positions of the Landau level crossing by varying the Ising-type SOC strength. I fitted the model on the experimentally determined Landau level positions and obtained the Ising-type SOC strength [T2]. Furthermore, I also modeled a WSe_2/BLG/WSe_2 heterostructure, where the Ising-type SOC is the opposite of the two graphene layers [T3]. I found that, by increasing the electric field, the SOC-induced gap closes and reopens, and the closing point could be used to obtain the Ising-type SOC strength [T3].

4. **I showed that the mobility of charge carriers in high-mobility single-layer graphene decreases with pressure due to the increased short-range and long-range scattering and also the increased effect of remote interfacial phonon coupling with hydrostatic pressure.** From field effect and Shubnikov-de Haas oscillations measurements, I showed that in high-mobility devices, the long-range scattering is the main source of the resistance and it remains dominant under pressure. From weak localization measurements, I found that in high-mobility devices, the short-range scattering is mainly caused by the edges of the sample which is insensitive to the pressure. I showed with in-plane magnetic field-dependent weak localization measurements, that the volume of the corrugations increased by increasing the pressure. From field effect measurements, I showed the increase of short-range and long-range scattering with pressure. From temperature-dependent magnetic focusing experiments, I

found that the pressure has a negligible effect on the combined effect of the electron-electron interactions and the acoustic phonon-electron coupling. From temperature-dependent field-effect transport measurements, I showed that the remote interfacial phonon-electron coupling increases with pressure. [T4]

Publications related to thesis points

T1 Bálint Szentpéteri, Peter Rickhaus, Folkert K. de Vries, Albin Márffy, Bálint Fülöp, Endre Tóvári, Kenji Watanabe, Takashi Taniguchi, Andor Kormányos, Szabolcs Csonka and Péter Makk, Tailoring the band structure of twisted double bilayer graphene with pressure. *Nano Letters* **21**(20), 8777–8784 (2021)

T2 Bálint Szentpéteri, Albin Márffy, Máté Kedves, Endre Tóvári, Bálint Fülöp, István Kükemezey, András Magyarkuti, Kenji Watanabe, Takashi Taniguchi, Szabolcs Csonka and Péter Makk, Increasing the proximity-induced spin-orbit coupling in bilayer graphene/WSe_2 heterostructures with pressure. Accepted in Physical Review B, arXiv:2409.20062

T3 Máté Kedves, Bálint Szentpéteri, Albin Márffy, Endre Tóvári, Nikos Papadopoulos, Prasanna K. Rout, Kenji Watanabe, Takashi Taniguchi, Srijit Goswami, Szabolcs Csonka and Péter Makk, Stabilizing the Inverted Phase of a WSe_2/BLG/WSe_2 Heterostructure via Hydrostatic Pressure. *Nano Letters* **23**(20), 9508–9514 (2023)

T4 Bálint Szentpéteri *et al.* Origin of the reduced charge carrier mobility in graphene under pressure. Manuscript in preparation.

Other publications

T5 Zoltán Kovács-Krausz, Anamul Md Hoque, Péter Makk, Bálint Szentpéteri, Mátyás Kocsis, Bálint Fülöp, Michael Vasilievich Yakushev, Tatyana Vladimirovna Kuznetsova, Oleg Evgenevich Tereshchenko, Konstantin Aleksandrovich Kokh, István Endre Lukács, Takashi Taniguchi, Kenji Watanabe, Saroj Prasad Dash, and Szabolcs Csonka, Electrically Controlled Spin Injection from Giant Rashba Spin–Orbit Conductor BiTeBr. *Nano Letters* **20**(7), 4782–4791 (2020)

T6 Bálint Fülöp, Albin Márffy, Simon Zihlmann, Martin Gmitra, Endre Tóvári, Bálint Szentpéteri, Máté Kedves, Kenji Watanabe, Takashi Taniguchi, Jaroslav Fabian, Christian Schönenberger, Péter Makk, and Szabolcs Csonka, Boosting

proximity spin–orbit coupling in graphene/WSe_2 heterostructures via hydrostatic pressure. *npj 2D Materials and Applications* **5**, 82 (2021)

References

[1] Valasa, S., Kotha, V. R. & Vadthiya, N. Beyond moore's law – a critical review of advancements in negative capacitance field effect transistors: A revolution in next-generation electronics. *Materials Science in Semiconductor Processing* **173**, 108116 (2024).

[2] Yoo, J., Nam, C.-Y. & Bussmann, E. Atomic precision processing of two-dimensional materials for next-generation microelectronics. *ACS Nano* **18**, 21614–21622 (2024).

[3] Novoselov, K. S. *et al.* Two-dimensional atomic crystals. *Proceedings of the National Academy of Sciences* **102**, 10451–10453 (2005).

[4] Novoselov, K. S. *et al.* Electric field effect in atomically thin carbon films. *Science* **306**, 666–669 (2004).

[5] Geim, A. K. & Grigorieva, I. V. Van der waals heterostructures. *Nature* **499**, 419–425 (2013).

[6] Qian, X., Liu, J., Fu, L. & Li, J. Quantum spin hall effect in two-dimensional transition metal dichalcogenides. *Science* **346**, 1344–1347 (2014).

[7] Ajayan, P., Kim, P. & Banerjee, K. Two-dimensional van der waals materials. *Physics Today* **69**, 38–44 (2016).

[8] Ferrari, A. C. *et al.* Science and technology roadmap for graphene, related two-dimensional crystals, and hybrid systems. *Nanoscale* **7**, 4598–4810 (2015).

[9] Bian, Z., Miao, J., Zhao, Y. & Chai, Y. Strong interlayer interaction for engineering two-dimensional materials. *Accounts of Materials Research* **3**, 1220–1231 (2022).

[10] Dean, C. R. *et al.* Boron nitride substrates for high-quality graphene electronics. *Nature Nanotechnology* **5**, 722–726 (2010).

[11] Yankowitz, M., Ma, Q., Jarillo-Herrero, P. & LeRoy, B. J. van der waals heterostructures combining graphene and hexagonal boron nitride. *Nature Reviews Physics* **1**, 112–125 (2019).

[12] Hennighausen, Z. & Kar, S. Twistronics: a turning point in 2d quantum materials. *Electronic Structure* **3**, 014004 (2021).

[13] Cao, Y. *et al.* Correlated insulator behaviour at half-filling in magic-angle graphene superlattices. *Nature* **556**, 80–84 (2018).

[14] Cao, Y. *et al.* Unconventional superconductivity in magic-angle graphene superlattices. *Nature* **556**, 43–50 (2018).

[15] Shah, S. J. *et al.* Progress and prospects of moiré superlattices in twisted tmd heterostructures. *Nano Research* **17**, 10134–10161 (2024).

[16] Fabian, J., Matos-Abiague, A., Ertler, C., Stano, P. & Zutic, I. Semiconductor spintronics. *Acta Physica Slovaca* **57**, 565–907 (2007).

[17] Ralph, D. & Stiles, M. Spin transfer torques. *Journal of Magnetism and Magnetic Materials* **320**, 1190–1216 (2008).

[18] Seneor, P. *et al.* Spintronics with graphene. *MRS Bulletin* **37**, 1245–1254 (2012).

[19] Han, W., Kawakami, R. K., Gmitra, M. & Fabian, J. Graphene spintronics. *Nature Nanotechnology* **9**, 794–807 (2014).

[20] Gmitra, M. & Fabian, J. Graphene on transition-metal dichalcogenides: A platform for proximity spin-orbit physics and optospintronics. *Physical Review B* **92**, 155403 (2015).

[21] Yan, W. *et al.* A two-dimensional spin field-effect switch. *Nature Communications* **7**, 13372 (2016).

[22] Yang, B. *et al.* Tunable spin–orbit coupling and symmetry-protected edge states in graphene/WS2. *2D Materials* **3**, 031012 (2016).

[23] Dankert, A. & Dash, S. P. Electrical gate control of spin current in van der waals heterostructures at room temperature. *Nature Communications* **8**, 16093 (2017).

[24] Omar, S. & van Wees, B. J. Spin transport in high-mobility graphene on WS2 substrate with electric-field tunable proximity spin-orbit interaction. *Physical Review B* **97**, 045414 (2018).

[25] Benitez, L. A. *et al.* Tunable room-temperature spin galvanic and spin hall effects in van der waals heterostructures. *Nature Materials* **19**, 170–175 (2020).

[26] Herling, F. *et al.* Gate tunability of highly efficient spin-to-charge conversion by spin hall effect in graphene proximitized with wse2. *APL Materials* **8**, 071103 (2020).

[27] Safeer, C. K. *et al.* Room-temperature spin hall effect in graphene/MoS2 van der waals heterostructures. *Nano Letters* **19**, 1074–1082 (2019).

[28] Yang, H. *et al.* Twist-angle-tunable spin texture in wse2/graphene van der waals heterostructures. *Nature Materials* (2024).

[29] Pimenta Martins, L. G. *et al.* High-pressure studies of atomically thin van der waals materials. *Applied Physics Reviews* **10** (2023).

[30] Maurand, R. *et al.* Fabrication of ballistic suspended graphene with local-gating. *Carbon* **79**, 486–492 (2014).

[31] Tóvári, E., Csontos, M., Kriváchy, T., Fürjes, P. & Csonka, S. Characterization of sio2/sinx gate insulators for graphene based nanoelectromechanical systems. *Applied Physics Letters* **105** (2014).

[32] Rickhaus, P. *et al.* Snake trajectories in ultraclean graphene p–n junctions. *Nature Communications* **6** (2015).

[33] Tóvári, E. *et al.* Gate-controlled conductance enhancement from quantum hall channels along graphene p–n junctions. *Nanoscale* **8**, 19910–19916 (2016).

[34] Tóvári, E., Makk, P., Rickhaus, P., Schönenberger, C. & Csonka, S. Signatures of single quantum dots in graphene nanoribbons within the quantum hall regime. *Nanoscale* **8**, 11480–11486 (2016).

[35] Fülöp, B. *et al.* New method of transport measurements on van der waals heterostructures under pressure. *Journal of Applied Physics* **130**, 064303 (2021).

[36] Kovács-Krausz, Z. *et al.* Signature of pressure-induced topological phase transition in zrte5. *npj Quantum Materials* **9** (2024).

[37] Yankowitz, M. *et al.* Dynamic band-structure tuning of graphene moiré superlattices with pressure. *Nature* **557**, 404–408 (2018).

[38] Yankowitz, M. *et al.* Tuning superconductivity in twisted bilayer graphene. *Science* **363**, 1059–1064 (2019).

[39] Song, T. *et al.* Switching 2d magnetic states via pressure tuning of layer stacking. *Nature Materials* **18**, 1298–1302 (2019).

[40] Chakraborti, H. *et al.* Electron wave and quantum optics in graphene. *Journal of Physics: Condensed Matter* **36**, 393001 (2024).

[41] Andrei, E. Y. & MacDonald, A. H. Graphene bilayers with a twist. *Nature Materials* **19**, 1265–1275 (2020).

[42] Bolotin, K. I., Ghahari, F., Shulman, M. D., Stormer, H. L. & Kim, P. Observation of the fractional quantum hall effect in graphene. *Nature* **462**, 196–199 (2009).

[43] Crossno, J. *et al.* Observation of the dirac fluid and the breakdown of the wiedemann-franz law in graphene. *Science* **351**, 1058–1061 (2016).

[44] Bandurin, D. A. *et al.* Negative local resistance caused by viscous electron backflow in graphene. *Science* **351**, 1055–1058 (2016).

[45] Wang, L. *et al.* Negligible environmental sensitivity of graphene in a hexagonal boron nitride/graphene/h-bn sandwich structure. *ACS Nano* **6**, 9314–9319 (2012).

[46] Wang, L. *et al.* One-dimensional electrical contact to a two-dimensional material. *Science* **342**, 614–617 (2013).

[47] Roche, S. *et al.* Graphene spintronics: the european flagship perspective. *2D Materials* **2**, 030202 (2015).

[48] Gmitra, M. & Fabian, J. Proximity effects in bilayer graphene on monolayer WSe2 : Field-effect spin valley locking, spin-orbit valve, and spin transistor. *Physical Review Letters* **119**, 146401 (2017).

[49] Wang, Z. *et al.* Strong interface-induced spin–orbit interaction in graphene on WS2. *Nature Communications* **6**, 8339 (2015).

[50] Wang, Z. *et al.* Origin and magnitude of 'designer' spin-orbit interaction in graphene on semiconducting transition metal dichalcogenides. *Physical Review X* **6**, 041020 (2016).

[51] Yang, B. *et al.* Strong electron-hole symmetric rashba spin-orbit coupling in graphene/monolayer transition metal dichalcogenide heterostructures. *Physical Review B* **96**, 041409 (2017).

[52] Wakamura, T. *et al.* Strong anisotropic spin-orbit interaction induced in graphene by monolayer ws2. *Physical Review Letters* **120**, 106802 (2018).

[53] Zihlmann, S. *et al.* Large spin relaxation anisotropy and valley-zeeman spin-orbit coupling in wse2/graphene/h-bn heterostructures. *Physical Review B* **97**, 075434 (2018).

[54] Amann, J. *et al.* Counterintuitive gate dependence of weak antilocalization in bilayer graphene/wse2 heterostructures. *Physical Review B* **105**, 115425 (2022).

[55] Afzal, A. M. *et al.* Gate modulation of the spin-orbit interaction in bilayer graphene encapsulated by ws2 films. *Scientific Reports* **8**, 3412 (2018).

[56] Tiwari, P. *et al.* Experimental observation of spin-split energy dispersion in high-mobility single-layer graphene/wse2 heterostructures. *npj 2D Materials and Applications* **6**, 68 (2022).

[57] Rao, Q. *et al.* Ballistic transport spectroscopy of spin-orbit-coupled bands in monolayer graphene on wse2. *Nature Communications* **14**, 6124 (2023).

[58] Omar, S. & van Wees, B. J. Graphene-ws2 heterostructures for tunable spin injection and spin transport. *Physical Review B* **95**, 081404 (2017).

[59] Avsar, A. *et al.* Spin–orbit proximity effect in graphene. *Nature Communications* **5**, 4875 (2014).

[60] Benítez, L. A. *et al.* Strongly anisotropic spin relaxation in graphene–transition metal dichalcogenide heterostructures at room temperature. *Nature Physics* **14**, 303–308 (2017).

[61] Ghiasi, T. S., Ingla-Aynés, J., Kaverzin, A. A. & van Wees, B. J. Large proximity-induced spin lifetime anisotropy in transition-metal dichalcogenide/graphene heterostructures. *Nano Letters* **17**, 7528–7532 (2017).

[62] Sun, L. *et al.* Determining spin-orbit coupling in graphene by quasiparticle interference imaging. *Nature Communications* **14**, 3771 (2023).

[63] Burg, G. W. *et al.* Correlated insulating states in twisted double bilayer graphene. *Physical Review Letters* **123**, 197702 (2019).

[64] Cao, Y. *et al.* Tunable correlated states and spin-polarized phases in twisted bilayer–bilayer graphene. *Nature* **583**, 215–220 (2020).

[65] He, M. *et al.* Symmetry breaking in twisted double bilayer graphene. *Nature Physics* **17**, 26–30 (2020).

[66] Rickhaus, P. *et al.* Correlated electron-hole state in twisted double-bilayer graphene. *Science* **373**, 1257–1260 (2021).

[67] Wang, Y. *et al.* Emergent symmetry and valley chern insulator in twisted double-bilayer graphene. *Physical Review Letters* **133**, 246401 (2024).

[68] Liu, X. *et al.* Tunable spin-polarized correlated states in twisted double bilayer graphene. *Nature* **583**, 221–225 (2020).

[69] Shen, C. *et al.* Correlated states in twisted double bilayer graphene. *Nature Physics* **16**, 520–525 (2020).

[70] Liu, L. *et al.* Isospin competitions and valley polarized correlated insulators in twisted double bilayer graphene. *Nature Communications* **13** (2022).

[71] Kuiri, M. *et al.* Spontaneous time-reversal symmetry breaking in twisted double bilayer graphene. *Nature Communications* **13** (2022).